Supporting Information

Long term highly reversible zinc metal anode regulated by polar molecular interface adsorption

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Experimental Section

Electrolytes and Cathode Materials Preparation. Methyl acetate (MA, Aladdin, AR) was added to deionised water to create the new hybrid solvents. The volume ratios of MA to water were set at 0:100, 16:84, 20:80, and 24:86, respectively, and can be represented as M0, M16, and M24. The $Zn(CF_3SO_3)_2$ ($Zn(OTf)_2$, Aladdin, AR) concentration was 1 mol L^{-1}. As previously reported, the $K_{1.15}V_5O_{13}\cdot 1.3H_2O$ (KVO) cathode materials were prepared. The working electrodes were prepared as follows: the as-prepared KVO was mixed with carbon black (acetylene black, AB) and polymer binder (polytetrafluoroethylene, PTFE) in a weight ratio of 7:2:1 with the help of ethanol. After drying, the mixture was pressed into a film and cut into disks. The mass of the cathode was measured by an electronic balance. Several films were weighed and then used as the cathode. The KVO mass loading is 10-15 mg cm^{-2}.

Materials Characterizations. The contact angles of Zn foils with different electrolytes were measured by the static contact angle measurements (Chengde Dingsheng JY-82C Video Contact Angle Tester). The NMR spectra of electrolytes were acquired with the AVANCE III 600MHz (Bruker). FTIR spectra were collected by using a FTIR spectrometer (Thermo Scientific Nicolet iS20). Microstructures of Zn anodes were investigated by using SEM (TESCAN MIRA3 LMH). The chemical compositions and crystal structures of cycled Zn foils were examined by XRD (Bruker D8, Cu Kα) and the chemical state of cycled Zn anodes surface was probed by XPS (Thermo Scientific

K-Alpha). The surface roughness curve of cycled Zn anodes was measured by AFM (Bruker Dimension ICON).

Electrochemical Characterization. The Zn||Zn symmetric cells, Cu||Zn asymmetric cells and KVO||Zn full cells were assembled into CR2032 coin cells. The cells were assembled by using 16 mm Zn plates (100 μm) as the anode and glass fiber as the separator. The electrochemical performance testing was conducted on a LAND CT2001 battery testing system. Electrochemical characterizations, such as LSV, Tafel and CA et al., were performed on the IviumSoft electrochemical workstation, where Zn foil, Pt foil and Ag/AgCl were used as the working, counter and reference electrodes, respectively. The LSV profiles were tested at 5 mV s^{-1}. The Tafel curves were measured from -1.1 to -0.8 V with a scan rate of 10 mV s^{-1}. The CA curves were performed at a constant potential of -0.15 V. Electrochemical impedance spectroscopy was tested within a frequency range of 100 kHz to 0.1 Hz. Cyclic voltammetry (CV) measurements were conducted in the voltage range of -0.2 V~0.5 V for Zn||Ti cells, -0.015 V~0.015 V for Zn||Zn cells. The activation energy (E_a) was analyzed by electrochemical impedance spectrum (EIS) tests under different temperatures. According to Arrhenius equation:

$$R_{ct}^{-1} = A\, e^{-\frac{E_a}{RT}} \tag{1}$$

in which A represents a pre-exponential factor, T is the absolute temperature (K), R_{ct} and R delegate the charge transfer resistance and standard gas constant (8.314 J mol^{-1} K^{-1}), respectively. the activation energy (E_a) was analyzed by electrochemical impedance spectrum (EIS) tests under different temperatures.

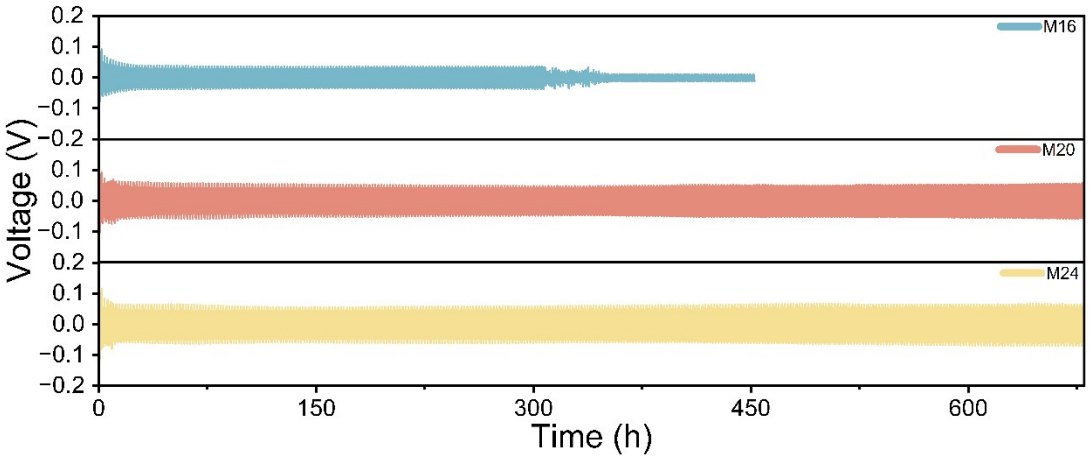

Figure S1. Cycling performance of Zn||Zn symmetrical cells with different electrolytes at 5 mA cm^{-2}, 5 mAh cm^{-2}.

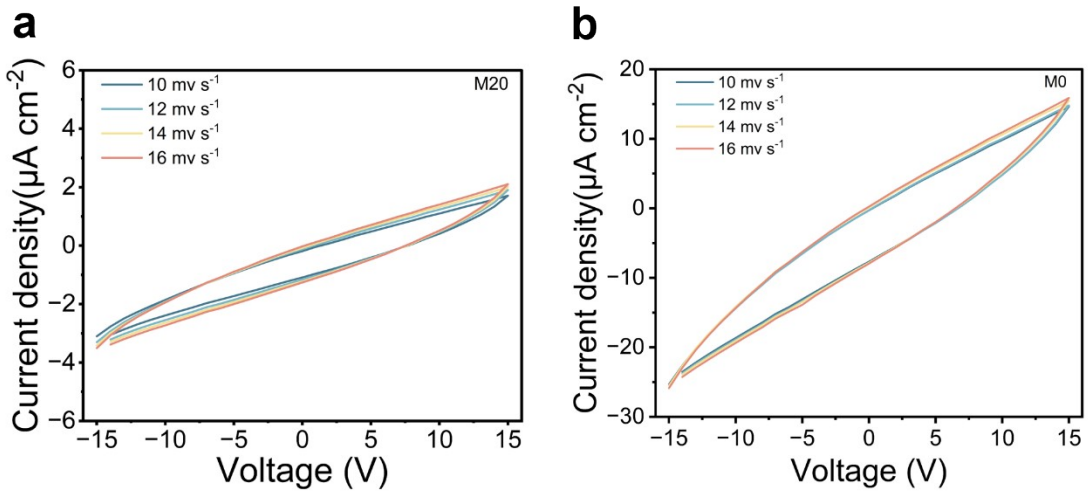

Figure S2 CV curves for EDL measurements in (a)M20 electrolyte, (b) M0 electrolyte.

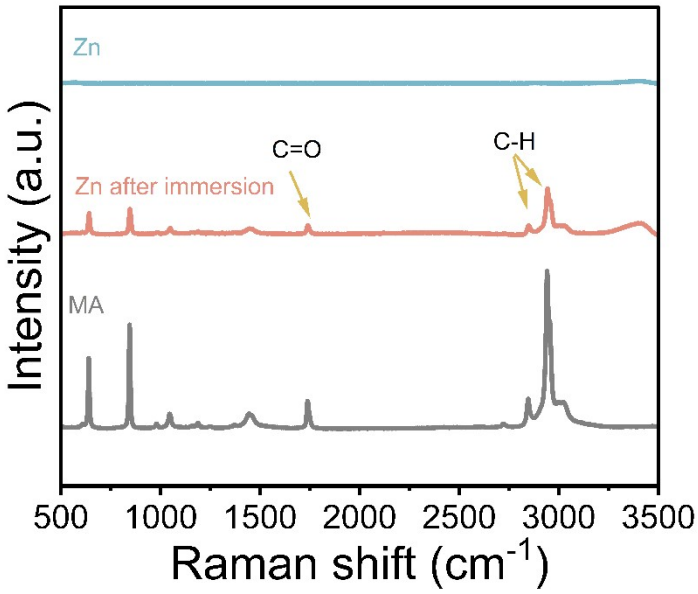

Figure S3 Raman spectra of Zn foil after soaking in M20.

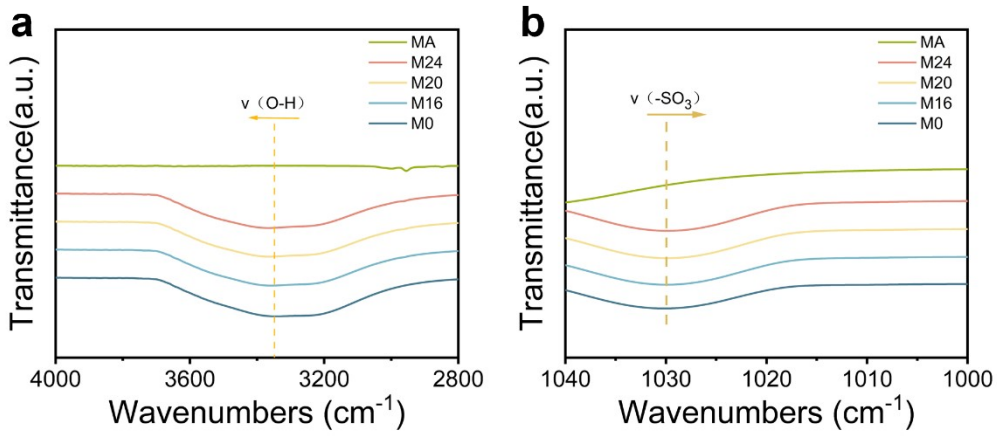

Figure S4 FTIR spectra of various electrolytes (a) O–H stretching vibration range, (b) $–SO_3$ vibration range.

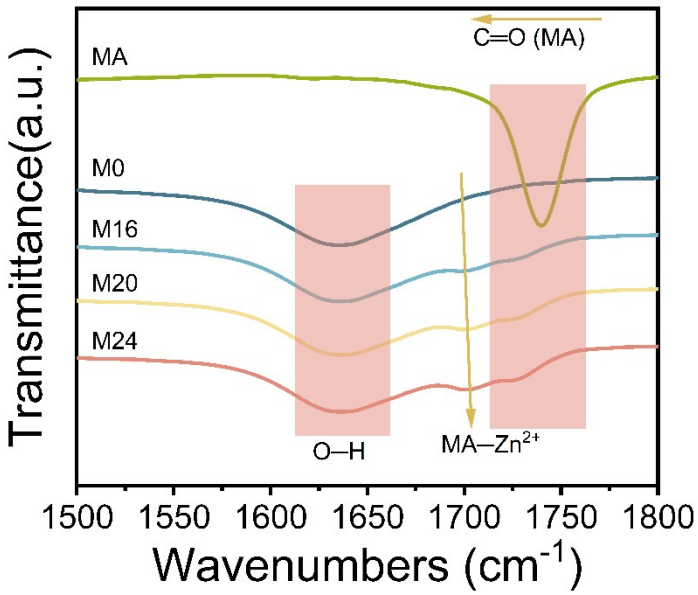

Figure S5 FTIR spectra of various electrolytes C=O vibration range.

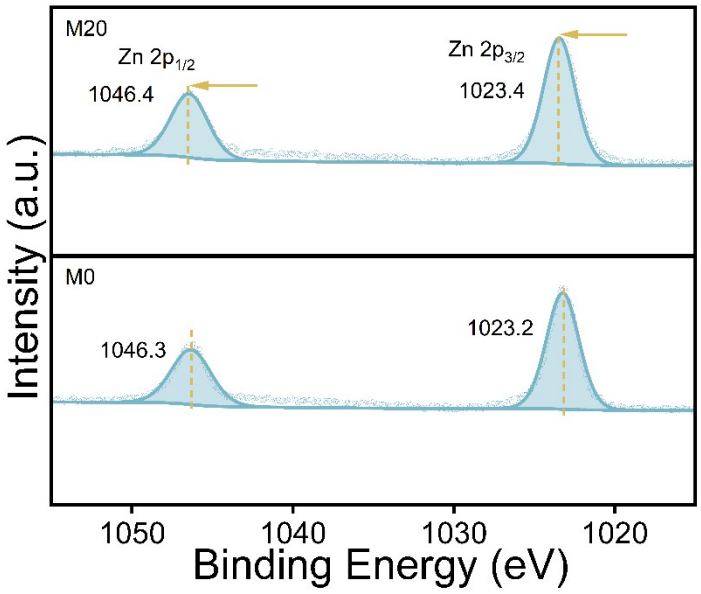

Figure S6 XPS spectrum after Zn foil immersion

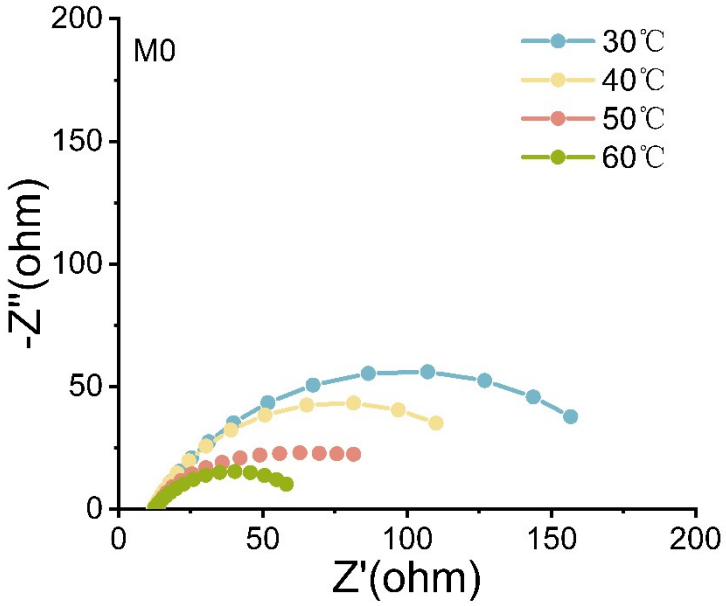

Figure S7 Nyquist plots of Zn||Zn cells tests at the temperature range of 30 to 60 ℃ with the M0 electrolyte.

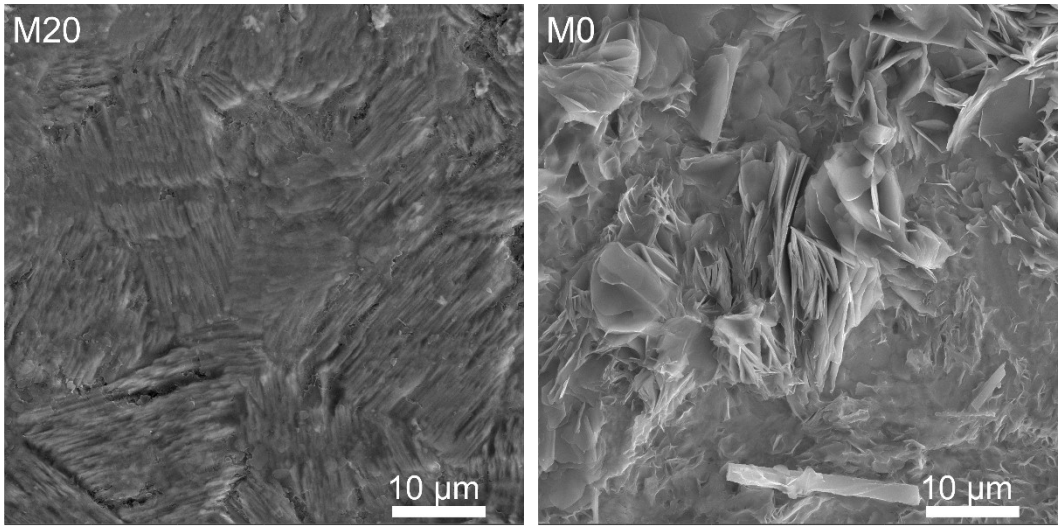

Figure S8 SEM images of Zn deposition after 1 h at 5 mA cm^{-2} in different electrolytes.

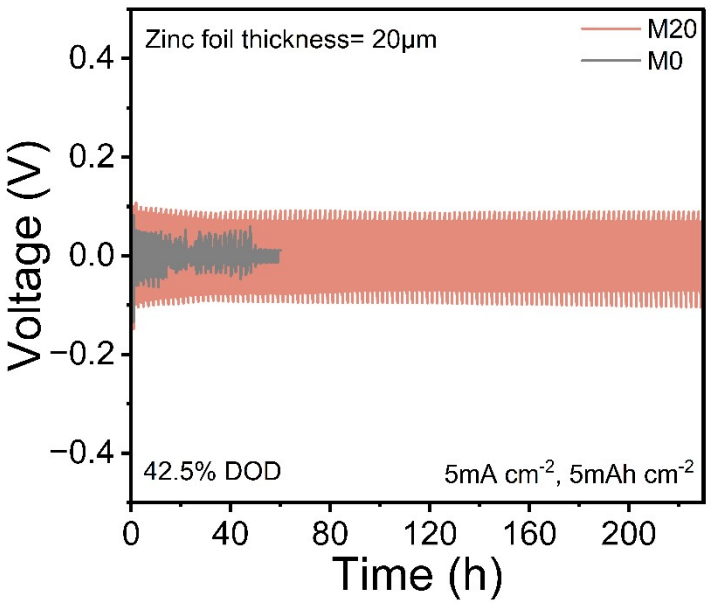

Figure S9 Cycling performance of Zn anodes at a DOD of 42.5%.

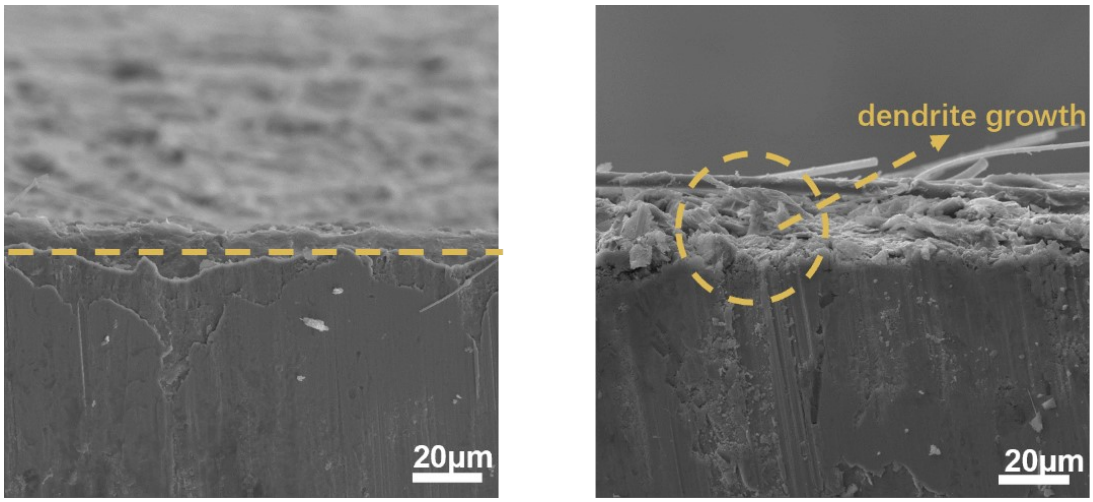

Figure S10 cross-sectional SEM images of cycled Zn anodes.

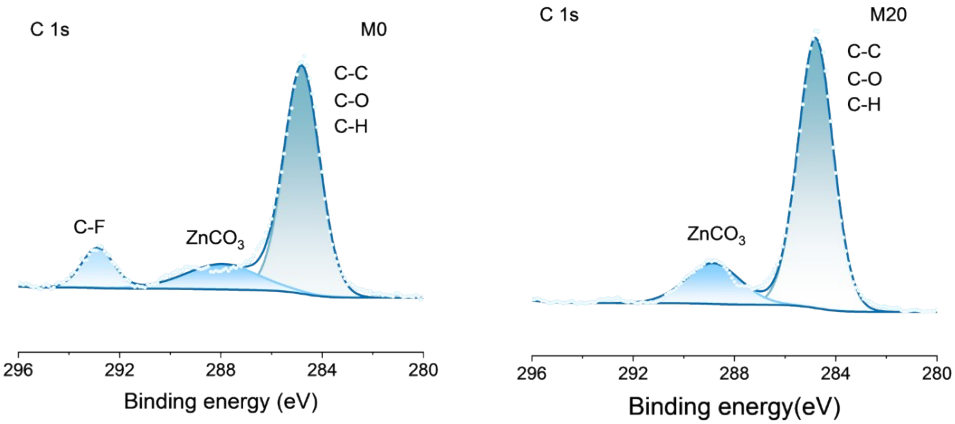

Figure S11 C 1s spectra of the XPS: (a) M0, (b) M20.

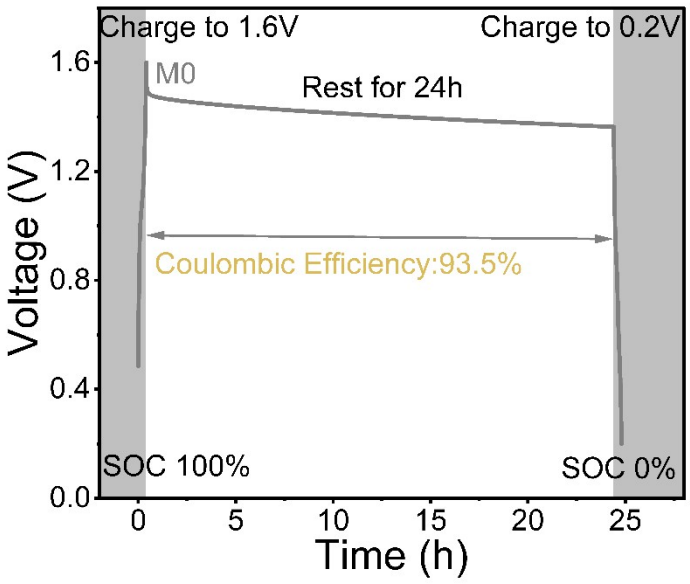

Figure S12 Self-discharge curves of M0.